

# Complementary Pu Resuspension Study at Palomares, Spain

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**COMPLEMENTARY Pu RESUSPENSION STUDY  
AT PALOMARES, SPAIN**

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## 1. INTRODUCTION

Soil in an area near Palomares, Spain, was contaminated with plutonium as a result of a mid-air collision of U.S. military aircraft in January 1966. The assessment for potential inhalation dose can be found in Iranzo et al., (1987). Long-term monitoring has been used to evaluate remedial actions (Iranzo et al., 1988) and there are many supporting studies of the Pu contamination at Palomares that have been carried out by the Centro de Investigaciones Energeticas, Medioambientales y Tecnologicas (CIEMAT) in Madrid.

The purpose of this study is to evaluate the resuspension of Pu from the soil in terms of Pu-concentrations in air and resuspension rates in a complementary investigation to those of CIEMAT but in an intensive short-term field effort.

It has long been recognized that the most significant pathway for human exposure to Pu is the inhalation of aerosolized-Pu attached to soil, and methods have been developed to estimate human exposure by this pathway (e.g. Anspaugh, Shinn, et al., 1975). From the perspective of health risk assessment, there are two physical processes that should be predicted: The process that produces the “respirable” Pu-concentration in air, and the process that causes a Pu flux into the air (rate of Pu-aerosol emissions) for subsequent redistribution. The term “respirable” is defined here as particles that are less than 10  $\mu\text{m}$  in aerodynamic diameter. Resuspended soil particles normally have a peak in the mass size-distribution at the mid-range between 1  $\mu\text{m}$  and 10  $\mu\text{m}$ .

A simple model for predicting the Pu concentration in air is the *resuspension factor* approach. In this concept, the Pu concentration in air is integrated over a period of at least several days to eliminate the variations due to wind and weather conditions, and these Pu concentrations are normalized by dividing the observed concentrations in air ( $C$ ,  $\text{Bq/m}^3$ ) by the local soil-Pu inventory from deposition ( $D$ ,  $\text{Bq/m}^2$ ). This method has proven useful around the world because once the resuspension factor,  $S_f$ , is estimated, then the concentration can be predicted from the deposition,  $D$ , that is,  $C = S_f D$ . The values,  $S_f$ , tend to a long-term limit between  $10^{-10} \text{ m}^{-1}$  and  $10^{-9} \text{ m}^{-1}$  (Garcia-Olivares and Iranzo, 1997).

This “steady-state” can be interrupted, however, by disturbances such as construction, traffic, etc. Another model, the *mass loading* approach, tries to deal with this problem by predicting the Pu-aerosol activity ( $A$ ,  $\text{Bq/g}$ ) and the suspended-particulate mass loading ( $M$ ,  $\text{g/m}^3$ ). The activity,  $A$ , would be predicted from an enhancement factor,  $E_f$ , and the surface soil activity,  $S_o$  ( $\text{Bq/g}$ ), that is,  $A = E_f S_o$ . In this model, the concentration can then be predicted by the product combination,  $C = E_f S_o M$ . Both  $S_o$  and  $M$  are easily measured. But both  $E_f$  and  $M$  can be expected to increase during disturbances. That  $E_f$  would increase with disturbance indicates that the Pu bindings with soil aggregates are somewhat fragile. In some cases  $M$  is predictable from dust emission factor models for various types of construction and agricultural activity. In studies performed over a wide number of Pu-contaminated sites, Shinn (1992) found that values of  $E_f$  were usually less than unity, typically 0.7, for the types of accidents such as at Palomares. For disturbances such as traffic, bulldozer blading, wildfire, and freezing-thawing cycles, Shinn reported that  $E_f$  values increased to between 2.5 and 6.5.

The second problem, predicting the Pu-aerosol emission rates, is determined by solving the flux equation  $F = K (dC/dz)$ , where  $K$  is conventionally measured as the turbulent diffusivity

for sensible heat, and the vertical gradient  $dC/dz$  is measured from vertically-spaced air samplers. Anspaugh, Shinn, *et al* (1975) simplified this even further by the approximation  $dC/dz = p C/z$  where  $p$  is the power-law parameter determined as the constant slope from the  $\log C$  versus  $\log z$  measurements.

The parameter  $p$  is a measure of the surface conditions and for suspended particulate mass loading has typical values of  $-0.2$  and a range between  $-0.05$  and  $-0.6$ . The negative sign indicates that suspended mass is decreasing with height in the air above the soil. The turbulent diffusivity  $K$  can be easily measured and varies directly with wind speed and height above ground. To determine the resuspension rate,  $R$ , the flux  $F$  ( $\text{Bq/m}^2 \text{ sec}$ ) is divided by the local soil Pu-inventory from deposition ( $D$ ,  $\text{Bq/m}^2$ ), that is  $R = F/D$ . This gives the fraction of the contamination being resuspended per second. Typical values for  $R$  are  $10^{-11} \text{ s}^{-1}$  to  $10^{-12} \text{ s}^{-1}$  (Shinn, *et al*, 1997, Garcia-Olivares and Iranzo, 1997). But in some cases soils have a higher  $R$ , e.g. more erodible, sandy soil or disturbed soil, and then local redistribution of Pu is a problem (Shinn *et al*, 1989).

The resuspension of Pu-particles from soil at Palomares has been monitored since the accident in 1966 and was summarized by Iranzo *et al* (1994). They reported that resuspension factors at Palomares indeed tend to the "steady-state" values between  $10^{-9} \text{ m}^{-1}$  and  $10^{-10} \text{ m}^{-1}$  within "several years" after the accident. Some disturbances due to construction of a reservoir pond near air sampling site 2-2, resulted in Pu concentrations in air that were increased during 1984-1987, but have returned to normal again. Iranzo *et al* (1987) showed that even with conservatively assumed annual exposures at the maximum observed Pu concentration in air, the calculated dose to the public was very low.

Nevertheless, continued monitoring and increased understanding of the Pu resuspension is a prudent means of maintaining control of public exposures.

This study complements the resuspension studies of CIEMAT at Palomares. It provides information not previously available on the observed mass loadings ( $M$ ), Pu-activity of aerosols ( $A$ ), enhancement factor ( $E_f$ ), daily fluctuations in the Pu concentration in air ( $C$ ), particle size distribution of the Pu in air in the respirable range, resuspension factor ( $S_f$ ), and resuspension rate ( $R$ ). These values are then compared with published data observed elsewhere.

## 2. METHODS

This is a brief summary of methodologies for field studies of resuspension. The detailed scope, responsibilities, and procedures were documented in a "Study Plan" that contains or refers to several different "Detailed Procedures", and is too long for this report. The air sampling procedures are described in Shinn (1995). The methods discussed here are the same, except for minor modifications as those reported in Shinn, Homan, and Robison (1997).

### 2.1 Air Sampling

Five types of air sampling were utilized by LLNL: (1) high-volume (HV) air samplers ( $100 \text{ m}^3/\text{h}$ ) for  $C$  and  $M$ , (2) cascade impactors ( $34 \text{ m}^3/\text{h}$ ) for  $C$  and  $M$  and the aerosol size distribution, (3) vertically-separated  $dC/dz$  samplers ( $100 \text{ m}^3/\text{h}$ ) by simultaneous HV at two

heights above the ground (1.1 m, and 2.1 m), (4) an ultra-high volume (UHV) air sampler that was changed daily (250 m<sup>3</sup>/h), and (5) a self-propelled isokinetic particle accumulator (IPA) with average rate of 7 m<sup>3</sup>/h. For each trial there was one UHV air sampler, one IPA, two day or night cascade impactors, two continuous cascade impactors, and two each vertically-separated pairs of HV. The IPA was an additional air sampler not used in our previous work. All together over 185,000 m<sup>3</sup> of air was sampled. Like filters were composited to reduce the number of samples analyzed to 28.

The IPA is commonly used in the Former Soviet Union and consists of a horizontal tube that pivots into the wind with a wind-driven propeller mounted at the tail to exhaust air through the tube, and a low resistance filter-impactor collection head on the inlet end. Tests with hot-wire anemometers showed that IPA was isokinetic (sampling speed equal to air speed) at wind speeds greater than its stall speed of 0.7 m/s.

Aerosols were collected on cellulose fiber media that were weighed and handled with rubber gloves to obtain high precision after the filter media were equilibrated to constant humidity and temperature. The face velocity of the samplers was sufficient to obtain better than 95% collection efficiency for solid particles. HV and cascade impactor air samplers were calibrated with critical orifice units before the field trials, and flow rates were monitored in the field. The UHV was calibrated after the field trials with a high volume laminar flow element, and flow rates were monitored with a pitot tube on a straight section of pipe placed in the exhaust duct. Minute-by-minute changes in the suspended mass loading, *M*, were monitored with a miniature, light-scattering nephelometer that was recorded with the weather data.

## **2.2 Analytical Methods**

For the transuranic isotopes, <sup>239-240</sup>Pu and <sup>241</sup>Am, special chemical methods were required. On filter media, the isotopes were recovered by acid total-dissolution, ion-exchange separation, and electrodeposition, and measured by alpha spectrometry using internal chemical yield tracers of <sup>242</sup>Pu and <sup>243</sup>Am. Details of the protocol are provided in Wong, *et al*, 1995. Quality assurance was carried out through adherence to established protocols, (Kehl, *et al*, 1995) and by use of quality control procedures (blank filters, control filters-carried to the site and back without use, and standards). Analytical accuracy has been maintained among these methods for the analysis of Pu (from atmospheric fallout) in shallow or deep marine sediments or in soil and sediment samples collected from the Pacific Test Sites at Bikini and Enewetak Atolls (close-in fallout). The method has also been used in the analysis of Pu in NIST Radioactivity Standard Reference Materials (Rocky Flats Soil and Columbia River Sediment) and in IAEA interlaboratory comparison samples. The alpha chemistry precision of the air filters was better than 0.005 Bq/ sample for <sup>239-240</sup>Pu and <sup>241</sup>Am.

## **2.3 Meteorological Methods**

Measurement of the turbulent diffusion characteristics and other relevant meteorological variables (for example, wind direction) was done with a special weather station set up in the field simultaneously with the air sampling. A meteorological method was used to determine

representativeness of the air samplers and meteorological sensors. At the typical height of 1.13 m in a short grass field, an air sampler is 90% representative of an upwind range (fetch) of 145 m independent of wind speed, (Shinn and Gouveia 1992).

An automatic (solar-charged) micrometeorological station was set up to continuously record wind speed, wind direction, and special variables that allow quality control of the estimate of turbulent diffusion. The special variables consisted of methods of estimation of the diffusivity of sensible heat through the energy balance technique (Fritschen-type net radiometers and soil heat-flux plates) and the Bowen Ratio technique, converted to diffusivity ( $K$ ) by means of measured air-temperature gradients. In addition, the sensible heat flux was measured independently by means of a one-dimensional (vertical) sonic anemometer with a fast-response thermocouple, using the eddy-correlation method. Comparison of the observed heat flux by the two techniques for optimal choice of coefficients constitutes the quality assurance of measured  $K$ . This  $K$  is used to estimate the Pu aerosol emission rates ( $F$ ), when combined with the measurement of the vertical gradient of radioactivity ( $dC/dz$ ) obtained by the vertical HV air samplers. The estimate is stability corrected by means of parameterization to the Monin-Obukov scaling factor with the choice of coefficients in the diabatic influence factors derived from the actual field data (Hanna, et al., 1982). The choice of coefficients does not have a particularly sensitive effect on  $K$ ; we found that a deviation of 50% from published, conventional values has only a 10% effect on estimation of  $K$  in typical field trials.

A battery-powered wind profile system with five, sensitive, cup anemometers was used to obtain aerodynamic roughness properties of the site for several successive 10-minute periods during strong winds and near-neutral stability conditions. These roughness properties (roughness length, 2 cm and zero-plane displacement, 10 cm) were used in the meteorological calculations of  $K$  and stability parameterizations discussed above.

### 3. RESULTS

Field trials were conducted in Palomares during June 1993 with logistical support provided by CIEMAT. The location of the field trials was at plot 2-0, generally downwind of plot 2-1, where resuspension studies were conducted by CIEMAT (Iranzo, et al., 1994). According to the representativeness criteria discussed above, air samples were integrated over at least 145 m. A heavy-duty generator was used to power the air samplers from June 19-30, except for a one-day generator failure on June 27. During the power failure the IPA air sampler continued to operate. The LLNL air samplers were located in a semi-circle within 20 m of the permanent CIEMAT meteorological tower and air samplers.

#### 3.1 Overview of the period, June 19-30, 1993

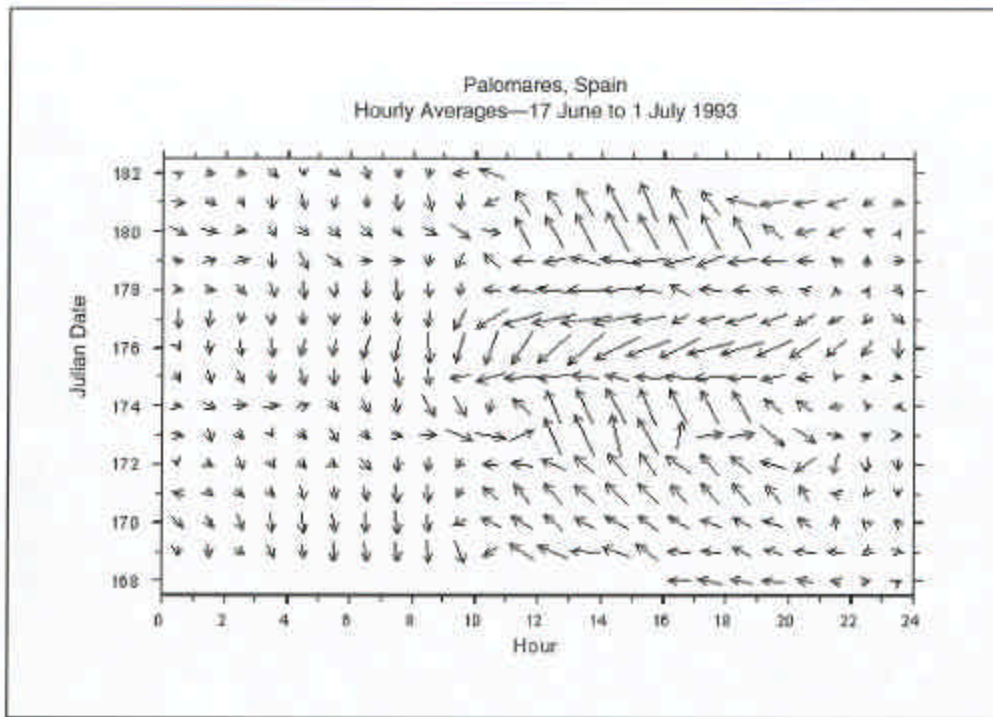
Weather conditions were sunny and clear during the entire period. Wind speeds were less than 1 m/s at night, and increased to a peak in the late afternoon. Peak speeds exceeded 3 m/s every day with peaks greater than 5 m/s on several days. The hourly-averaged wind vectors are shown in Fig 1 for each hour of the day over the 14 days. Vectors pointing toward the



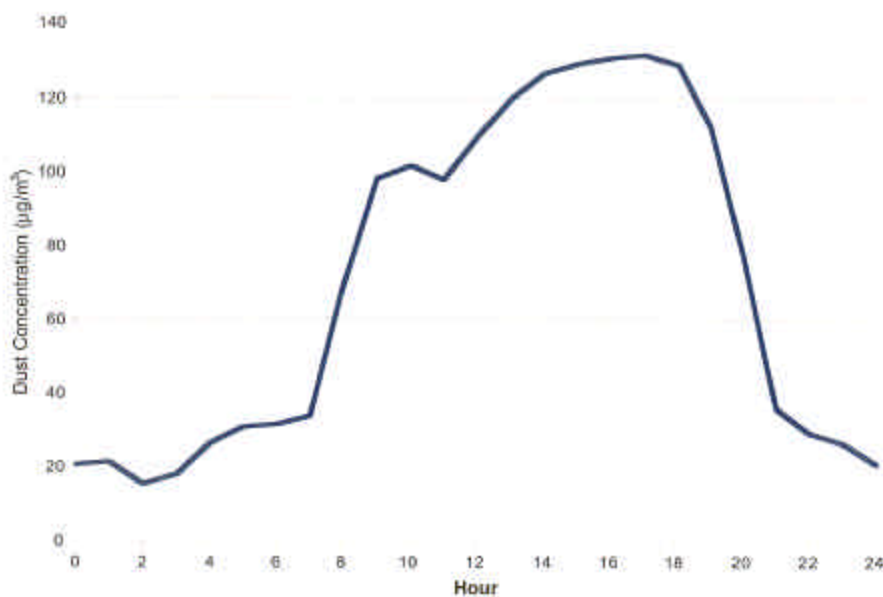
abscissa (downward as viewed) indicate winds from the north; vectors pointing to the left as viewed indicate winds from the east, etc. Vector length indicates mean speed.

Cascade impactors were used to compare the differences between night and day during June 19-30 (Julian days 170-181); daytime samples were collected during the period 10:20 h to 20:00 h (local time) and night samples were collected during the period 20:00 h to 10:20 h. The upper cutoff particle size of the impactors was 7  $\mu\text{m}$ . The impactor data showed that the night period had 15% lower Pu concentrations in air (C) compared to the 24-h average. Likewise, the daytime period had 22% higher C than the 24-h average. The weather data showed two distinct regimes between day and night (Fig 1). At night the winds were weak from the north or northwest (land drainage breeze) from about 20:00 h to 10:00 h, and during the day winds were stronger and from the northeast to southeast (sea breeze) from about 10:00 h to 20:00 h. This means that the sampling location was more representative of Plot 2-1. Dust loading, M, in the air was similar; there was about 20% lower M at night and about 30% higher M during daytime compared to the 24-h average. The average mass loading hour by hour during the day is also evident in typical data from the light-scattering nephelometer (Fig 2). The reason for the differences between night and day is mainly due to increased atmospheric diffusivity, K, during the day. Human exposures would be lower at night, partly because residents would be in their quarters, but also because of the reduced outdoor levels of Pu in the air.

The average level of Pu concentration in air (C) during the period 19-30 June downwind of plot 2-1 measured by the cascade impactors was  $8.5 \mu\text{Bq}/\text{m}^3$  with 42% relative variation (defined as the ratio of standard deviation to the mean) between samplers. This variation comes mostly from the variation detected in the aerosol activity. The mean aerosol activity (A) was  $0.12 \text{ Bq}/\text{g}$  with 41% relative variation. The mean mass loading, M, was  $70 \mu\text{g}/\text{m}^3$  with 7% relative variation (between 8 HV units). The observed value of C agrees with values reported by CIEMAT, considering the observed variation in A and the difference in location of instruments. These and other data are summarized in Table 1.



**Figure 1.** Hourly average wind vectors at Palomares Plot 2-1, each day during 17 June through 30 June, (Julian days 168-182) 1993. In this standard notation vectors as viewed pointing downward indicate winds from the north, vectors as viewed pointing left are from the east, etc. Vector lengths indicate mean speed.



**Figure 2.** Respirable dust suspended over the 24-hour period at Plot 2-1, Palomares, Spain, June 1993 (composite of data).

**Table 1.** Summary of resuspension and soil data of upwind Plot 2-1, June 19-30.

<b>M</b> <b>mg/m<sup>3</sup></b>	<b>C</b> <b>mBq/m<sup>3</sup></b>	<b>D</b> <b>kBq/m<sup>2</sup></b>	<b>S<sub>o</sub></b> <b>Bq/g</b>	<b>MAD</b> <b>mm</b>	<b>A</b> <b>Bq/g</b>	<b>E<sub>f</sub></b>	<b>S<sub>f</sub></b> <b>10<sup>-10</sup> m<sup>-1</sup></b>
70	8.5	35	0.44	3.7	0.12	0.28	2.4

The mass median aerodynamic diameter (MMAD) was 3.6  $\mu\text{m}$  and the activity median aerodynamic diameter (AMAD) was 3.3  $\mu\text{m}$  as determined from the cascade impactor data, but the variation between cascade impactors was larger than these differences. The data were thus combined to provide a median aerodynamic diameter (MAD) of 3.7 for both mass and activity. (See Table 1.) The standard deviation of these mean values was 0.9  $\mu\text{m}$ .

The particle size distributions were approximately lognormal with mass geometric standard deviation (MGSD) of 3.7 and an activity geometric standard deviation (AGSD) of 3.4. See Table 2. Again, these differences are not significant and data were combined to determine the mean GSD for both activity and mass of 3.5 with a standard deviation of 1.1. This means that the size distribution was very broad with a calculated 84th-percentile of 13  $\mu\text{m}$ . This kind of a distribution is commonly found in Pu-containing soil aerosols.

**Table 2.** Particle-size observations downwind of Plot 2-1, June 19-30.

<b>MMAD</b> <b>(Mass Median)</b> <b>mm</b>	<b>MGSD</b> <b>Mass Geometric</b> <b>Standard Dev.</b>	<b>AMAD</b> <b>(Activity Median)</b> <b>mm</b>	<b>AGSD</b> <b>Activ. Geometric</b> <b>Standard Dev.</b>
3.6	3.7	3.3	3.4

According to Iranzo, *et al* (1994) the soil activity to a depth of 5 cm is 0.44 Bq/g in Plot 2-1. Although the equipment was located on Plot 2-0, the strongest winds were from the direction of Plot 2-1. Using the soil density of 1600 Kg/m<sup>3</sup> the calculated soil inventory (D) is 35 kBq/m<sup>2</sup>. Thus we derive a resuspension factor (C/D) of  $2.4 \times 10^{-10} \text{ m}^{-1}$  that agrees very well with the values between  $10^{-10} \text{ m}^{-1}$  and  $10^{-9} \text{ m}^{-1}$  reported by them for nearby plot 2-2. The enhancement factor E<sub>f</sub> was 0.3 (the ratio of activity in respirable aerosols to the activity in soil), and is low compared to typical value of the 0.7 generally observed in undisturbed soil by Shinn (1992). Iranzo et al., (1994) define a similar coefficient, the dust loading factor, S<sub>e</sub>, that is equal to E<sub>f</sub>M, so we can estimate S<sub>e</sub> = (0.3)  $\times$  (70  $\mu\text{g}/\text{m}^3$ ) = 21  $\mu\text{g}/\text{m}^3$  from our data, that compares to 93  $\mu\text{g}/\text{m}^3$  which they report as an annual average for plot 2-2. It is quite possible that M is larger on the average at plot 2-2 than we observed for plot 2-1 in this June.

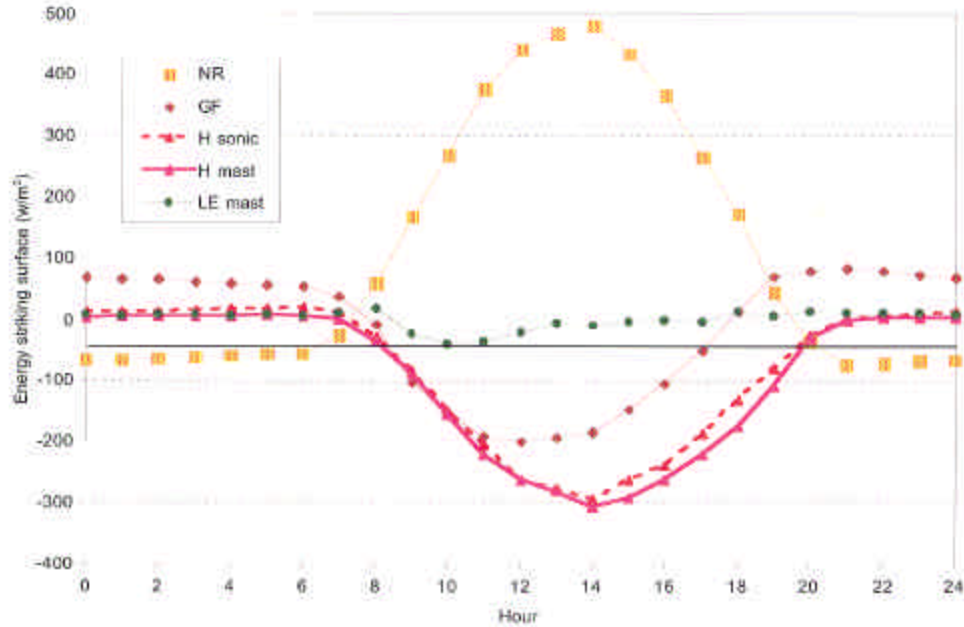
As an additional check of LLNL and CIEMAT analytic results, we compared data on the isotopic ratio  $^{239-240}\text{Pu}/^{241}\text{Am}$ . On four UHV filters we observed a mean Pu/Am ratio of 7.1 with a relative variation of 30%, which compares well with a mean value of 6.5 for nearby plot 2-2 reported by Iranzo, et al., (1986). We also observed a mean value of 0.020 for the ratio  $^{238}\text{Pu}/^{239-240}\text{Pu}$  with a relative variation of 15% on those filters.

UHV data show variations from day to day that were lognormally distributed in frequency with a geometric standard deviation of 4.9, meaning that the 98th-percentile would be expected to be 24 times the median. This C variation that is detectable only by the UHV samplers would be expected in other air samplers if we had the ability to detect Pu at much lower levels. The median value observed for the UHV was  $38 \mu\text{Bq}/\text{m}^3$ , which is much larger than the mean value of  $8.5 \mu\text{Bq}/\text{m}^3$  observed with other samplers. The explanation is that the UHV in this configuration samples particles that include the respirable size but also those much larger. So the UHV sampled a much larger population of suspended particulates.

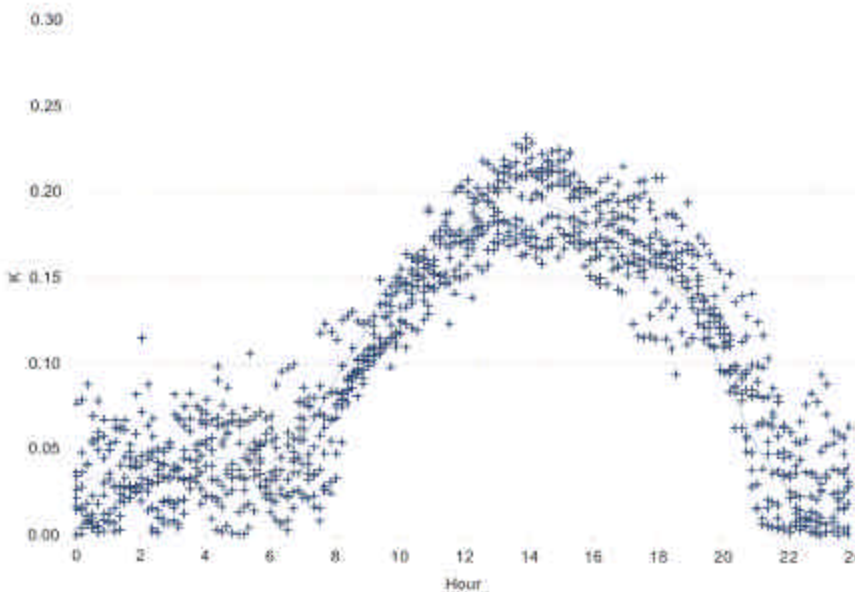
### **3.2 Special Trial Periods**

Two sequential field trials were conducted. Trial 1 was June 22-26, and Trial 2 was June 26-30. During these periods, the weather was nearly the same with the exception of June 25 of Trial 1 (Julian day 176) when the winds were strong with a northeasterly component during the day (see Fig 1). A composite of daily earth energy budget (Fig 3) showed strong solar energy input ( $\text{watts}/\text{m}^2$ ) in the form of net global radiation (NR), with high rates of ground heat flux (GF) and sensible heat flux (H), and almost no evaporation (LE). The error in this budget is about  $30 \text{ watts}/\text{m}^2$ , where heat gained by the surface is positive and heat lost from the surface is negative. The Bowen Ratio (H/LE) was greater than 30. The two methods for measuring sensible heat flux to determine diffusivity K, the sonic-anemometer eddy-correlation (H sonic) and the flux-gradient (H mast) were nearly identical as seen on Fig 3. Fig 4 shows the composite of K with its diurnal variations.

The measurement of the gradient  $dC/dz$ , expressed as the parameter p, showed a value of -0.11 with relative variation 29% in the first trial, and a value of -0.23 with relative variation 3.5% in the second trial. There is no apparent reason why they should be different for the two trials, but the values are comparable to those observed elsewhere. Using the average values of K at a height of 1.4 m, C measured by the HV at a height of 1.13 m, and p, we calculated a Pu-flux, F, and a resuspension rate, R, for both trials. See Table 3.



**Figure 3.** Earth surface energy budget (watts/m<sup>2</sup>) composited over several days, June 1993. **NR** is net global radiation, **GF** is ground heat flux, **LE** is evaporation, **H<sub>mast</sub>** is sensible heat flux by the gradient method, and **H<sub>sonic</sub>** is the sensible heat flux by the eddy-correlation method. Energy adding to the surface is positive and energy leaving the surface is negative.



**Figure 4.** Measured diffusivity for sensible heat,  $K$  (m<sup>2</sup>/s), used for calculating Pu-particle flux ( $F$ ) and resuspension rate ( $R$ ). All 10-minute data are shown, as measured, during the diurnal period.

**Table 3.** Resuspension data by trial period, downwind of Plot 2-1.

	K	C (HV)	C (UHV)	C (IPA)	p	F	R
June	m <sup>2</sup> /s	mBq/m <sup>3</sup>	mBq/m <sup>3</sup>	mBq/m <sup>3</sup>		mBq/m <sup>2</sup> s	10 <sup>-12</sup> s <sup>-1</sup>
22-26	0.11	8.1	59	-----	-0.11	0.07	2.0
26-30	0.10	8.5	11	9.6	-0.23	0.14	3.9

The values of R are in the same range,  $10^{-11} \text{ s}^{-1}$  to  $10^{-12} \text{ s}^{-1}$ , as observed by LLNL elsewhere (Shinn et al., 1997). This means that in spite of a dusty environment, the Pu is not redistributing at an unusually high rate. For example, if we determine the half-time for wind removal of Pu at this rate, it would be 10,500 y to 20,400 y.

The values of C obtained by the HV are largely confirmed by the UHV and IPA. The UHV values in Table 3 are medians of the samples that had large daily fluctuations, and were sampling larger particles as discussed previously. The IPA confirmed the values of C measured by the HV; the IPA sample was accidentally damaged in the first trial, but was useful in the second trial during the power failure on 27 June.

#### 4. CONCLUSIONS

This study complements the resuspension studies of CIEMAT at Palomares with additional information, and with confirmation of their previous studies. Observed mass loadings (M) were an average of  $70 \mu\text{g}/\text{m}^3$  with peaks in the daytime of  $130 \mu\text{g}/\text{m}^3$  and low values at night below  $30 \mu\text{g}/\text{m}^3$ . The Pu-activity of aerosols (A) downwind of plot 2-1 was  $0.12 \text{ Bq/g}$  and the enhancement factor ( $E_f$ ) had a value of 0.3, which is low but similar to a typical value of 0.7 for other undisturbed sites. This  $E_f$  value may increase further away from ground zero.

The particle size distribution of the Pu in air measured by cascade impactors was approximately lognormal with a median aerodynamic diameter of  $3.7 \mu\text{m}$  and a geometric standard deviation of 3.5 in the respirable range. This peak midway between  $1 \mu\text{m}$  and  $10 \mu\text{m}$  in the respirable range is commonly observed.

Daily fluctuations in the Pu concentration in air (C) detected by the UHV were lognormally distributed with a geometric standard deviation of 4.9 indicating that the 98th percentile would be 24 times as high as the median. Downwind of plot 2-1 the mean Pu concentration in air, C, was  $8.5 \mu\text{Bq}/\text{m}^3$ . The resuspension factor ( $S_f$ ) was  $2.4 \times 10^{-10} \text{ m}^{-1}$  and agrees very well with the values between  $10^{-10} \text{ m}^{-1}$  and  $10^{-9} \text{ m}^{-1}$  previously reported. We observed a mean Pu/Am ratio of 7.1 with a relative variation of 30%, which compares well with a mean value of 6.5 for nearby plot 2-2. The resuspension rate (R) was in the middle of the range,  $10^{-11} \text{ s}^{-1}$  to  $10^{-12} \text{ s}^{-1}$  as observed in other stable sites, and indicates low potential for Pu redistribution.

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## 6. REFERENCES

- Anspaugh, L.R., J. H. Shinn, P. L. Phelps, and N. C. Kennedy (1975), "Resuspension and Redistribution of Plutonium in Soils," *Health Physics*, **29**: 571–582.
- Garcia-Olivares, A. and C. E. Iranzo (1997), "Resuspension and Transport of Plutonium in the Palomares Area," *J. Environ. Radioactivity*, **37**(1): 101–114.
- Hanna, S. R., G. A. Briggs, and R. P. Hosker (1982), *Handbook on Atmospheric Diffusion*, U.S. Department of Energy, Technical Information Center, DOE/TIC 11223.
- Iranzo, C. E., A. Espinosa, and J. Martinez, 1994, Resuspension in the Palomares Area of Spain: A summary of experimental studies, *J. Aerosol Sci.*, **25**: 833–841.
- Iranzo, E., E. Mingarro, S. Salvador, C. E. Iranzo, and P. Rivas (1986), "Geochemical Distribution of Plutonium and Americium in Palomares Soil," in *The Cycling of Long-lived Radionuclides in the Biosphere: Observations and Models*, C. E. C., Brussels
- Iranzo, E., S. Salvador, and C. E. Iranzo (1987), "Air concentrations of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  and potential radiation doses to persons living near Pu contaminated areas in Palomares (Spain)," *Health Physics*, **52**:453–461.
- Iranzo, E., A. Espinosa, and C. E. Iranzo (1988), "Evaluation of remedial actions taken in a agricultural area contaminated by transuranides," in *The Impact of Nuclear Origin Accidents on Environment.*, C. E. C., Brussels.
- Kehl, S. R., M. E. Mount, and W. L. Robison (1995), *The Northern Marshall Islands Radiological Survey: A Quality Control Program for Radiochemical and Gamma Spectroscopy Analysis*, Technical Report UCRL-ID-120429, Lawrence Livermore National Laboratory, Livermore, CA.
- Shinn, J. H., E. H. Essington, F. L. Miller, Jr., T. P. O'Farrell, J. A. Orcutt, E. M. Romney, J. W. Shugart, and E. R. Sorom (1989), "Results of a Cleanup and Treatment Test at the Nevada Test Site: Evaluation of Vacuum Removal of Pu-Contaminated Soil," *Health Phys.*, **57**(5): 771–779.

Shinn, J. H. (1992), "Enhancement Factors for Resuspended Aerosol Radioactivity: Effects of Topsoil Disturbance," in *Proceedings of the Fifth International Conference on Precipitation Scavenging and Atmosphere-Surface Exchange Processes*, 3:1183–1193, S. E. Schwartz and W. G. N. Slinn, eds., Hemisphere Publishing Corp., Washington and Philadelphia.

Shinn, J. H. and F. J. Gouveia (1992), *The Footprint Area Influencing a High Volume Air Sampler*, Technical Report UCRL-ID-112181, Lawrence Livermore National Laboratory, Livermore, CA.

Shinn, J. H. (1995), *Protocols of Radiocontaminant Air Monitoring for Inhalation Exposure Estimates*, Technical Report UCRL-ID-122254, Lawrence Livermore National Laboratory, Livermore, CA.

Shinn, J. H., D. N. Homan, and W. L. Robison (1997), "Resuspension Studies in the Marshall Islands," *Health Physics*, **73**: 248–257.

Wong, K. M, T. A. Jokela, and V. E. Noshkin (1995), *Radiochemical Procedures for Analysis of Pu, Am, Cs, and Sr in Water, Soil, Sediments, and Biota Samples*, Technical Report UCRL-ID-116497, Lawrence Livermore National Laboratory, Livermore, CA.